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Weekly periodicities of Aerosol Optical Thickness over Central Europe – evidence of an anthropogenic direct aerosol effect

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Abstract. Statistical analyses of data from ground-based sun photometer stations in Central Europe are presented. All stations are part of the Aerosol Robotic Network (AERONET), and only data of the highest data quality level 2.0 has been applied. The averages by weekday of Aerosol Optical Thickness (AOT) at a wavelength of 440 nm of 12 of the 14 stations in the investigation area show a weekly periodicity with lowest values on Sunday and Monday, but greatest values from Wednesday until Saturday, that is significant at least on a 90% level. The stations in Germany and in Greater Paris show weekly cycles with ranges of about 20% on average. In Northern Italy and Switzerland this range is about 10% on average. By applying several checks, we exclude that the weekly cycles were caused by a maintenance effect or by different retrieval conditions as a consequence of a weekly cycle in cloud cover. The corresponding weekly cycle of anthropogenic gaseous and particulate emissions leads us to the conclusion of the anthropogenic origin of the weekly AOT cycle. Since these AOT patterns are derived from the reduction of the direct sun radiation by the columnar atmospheric aerosol, this result represents strong evidence for an anthropogenic direct aerosol effect on shortwave radiation. Furthermore, this study makes a first contribution to the understanding and explanation of recently observed weekly periodicities in meteorological variables as temperature in Germany.

1 Introduction

Despite the progress that recently has been made in climate research, the role of anthropogenic aerosol is still a key factor of uncertainty in understanding and quantifying the present and future global climate change (IPCC, 2007). Although the knowledge about the direct aerosol effect is better than the one about the indirect aerosol effect, solely the range of uncertainty of the anthropogenic direct effect is of the order of half the size of the total anthropogenic radiative forcing according to that report. The range of uncertainty of the anthropogenic indirect effect is even quantified as of the same order as the total anthropogenic radiative forcing, to some extent a consequence of its manifold manifestations (Lohmann and Feichter, 2005). Therefore, innovative methods to improve our knowledge about the atmospheric aerosol, its interactions, and its better representation in meteorological models are needed.

In the recent years, a series of studies reported weekly periodicities in various meteorological variables. Gordon (1994) found a significant but very small weekly temperature cycle for the northern hemisphere for the period 1979–1992. Simmonds and Keay (1997) published local weekly cycles in temperature and precipitation in Melbourne, Australia, for the years 1960–1994. Cerveny and Balling (1998) showed weekly periodicities of precipitation and tropical cyclone maximum wind speed over the North-West Atlantic region, and also the difference of day and night wind speed in tropical cyclones had a weekly periodicity (Cerveny and Balling, 2005). Forster and Solomon (2003) found a weekend effect in the daily temperature range in different regions of the world, and especially large scale patterns of areas with a positive or a negative weekend effect over the USA. Gong et al. (2006) reported increasing weekly cycles in various meteorological parameters including temperature and precipitation in most parts of China that depend on the season. Bäumer and Vogel (2007) found weekly periodicities in various meteorological parameters including temperature and precipitation in Germany in the period 1991–2005, and the temperature cycle with maxima on Wednesday and minima on Saturday also occurred at remote sites as on high mountains.

In some of these studies, the anthropogenic fraction of the atmospheric aerosol was made responsible for the weekly

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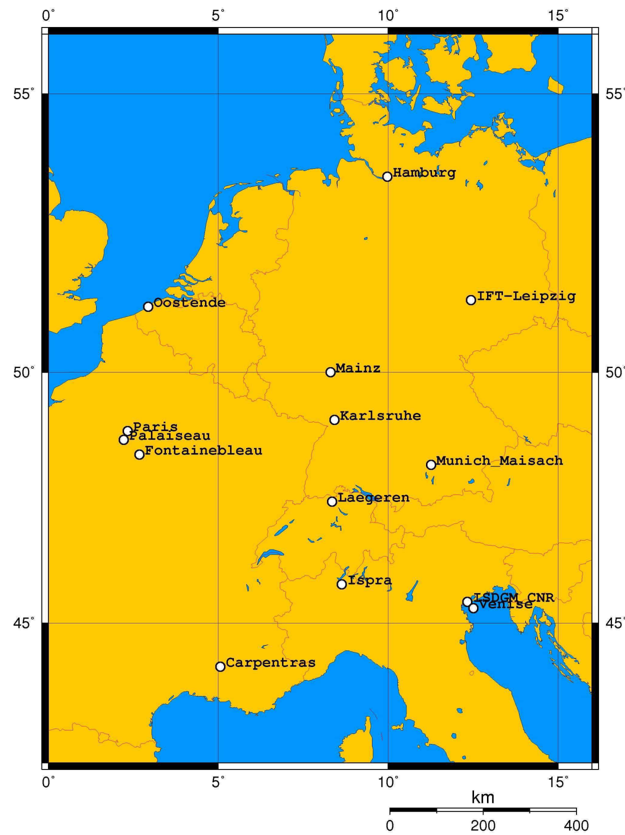

Fig. 1. Map showing positions of the AERONET sun photometer stations.

periodicities, but this hypothesis has not been proven yet in a satisfying way. In fact, there are weekly periodicities of near-surface aerosol properties at different locations as shown by e.g. Delene and Ogren (2002) and Shutters and Balling (2006). Differences of the visibility and the PM10 concentration between weekdays and weekends in Taiwan were published by Tsai (2005). With respect to column properties, Jin et al. (2005) reported a weekly cycle of aerosol optical thickness in New York, but only in summer and under further additional conditions that significantly reduced the size of the used data set. But still the question remains whether there is a weekly cycle of aerosol with a considerable vertical extent on larger horizontal scales than the urban scale. In the case of the trace gas NO_2 this could be shown by Beirle et al. (2003). They analyzed satellite data and demonstrated weekly cycles of tropospheric columnar NO_2 over many industrialized regions in the world including Germany and northern Italy. Ordonez et al. (2006) confirmed this finding for northern Italy both based on satellite measurements and derived from near-surface measurements.

In this paper, we investigate if there is a weekly periodicity in the columnar aerosol content over Central Europe. Therefore, we analyze time series of AERONET sun photometer aerosol optical thickness (AOT).

2 Data

Aerosol Robotic Network (AERONET) is a federated international network of sun and sky radiometers that exists since 1993. There are more than 300 automatically operating instruments worldwide (Holben et al., 1998; Holben et al., 2001) that have been used for a large number of applications. For instance, Eck et al. (1999) investigated the optical properties of biomass burning aerosol. Smirnov et al. (2002) found a diurnal variability of aerosol optical thickness at urban and industrial sites. Most of the stations showed an increase of the AOT by about 10% to 20% in the course of the day, predominantly as a response to anthropogenic emissions.

In this study, we have been testing the data sets measured at many AERONET sun photometer stations all over Central Europe for a weekly periodicity. We solely used data of the best data quality level 2.0 provided as daily averages. These data are cloud-screened and quality-assured to exclude a negative influence from insufficient cloud correction, especially as a weekly cycle in cloud cover as shown for Germany by Bäumer and Vogel (2007) might affect the retrieval of AOTs otherwise. This comprises several checks by AERONET as e.g. pre- and post-field calibration, instrument performance check, data consistency checks, and AOT spectral dependency check. Since the sun photometer carries out direct sun radiation measurements, there are no data available from completely overcast days. The list of applied stations in the investigation area, their coordinates, the respective according measurement period, and the number of measurement days is given in Table 1. Not all the stations measure AOT at a wavelength of 500 nm but at 440 nm. Hence we have chosen to solely show results for AOT at 440 nm in this work, but where available, the AOTs at 500 nm show the same pattern. The stations are situated in France, Belgium, Germany, Switzerland, and Italy. Figure 1 shows a map including the positions of the stations.

3 Results

In Table 2, an overview of the statistics for the applied AERONET stations is given. Average AOT440 absolute values vary from about 0.2 at Laegeren in Switzerland to 0.39 in Venice, Italy. The minimum between Saturday and Tuesday and the maximum between Wednesday and Saturday of the average weekday AOT440 could be proven as significantly different for 12 of the 14 stations at least on a 90% level by applying a t-test for the respective two weekdays.

Figure 2 shows the mean relative weekly variability of AOT at 440 nm for the 5 German AERONET stations and their average AOT at 440 nm that is computed as the relative percent departure from the mean value. Despite some variation from day to day and station to station, all stations have above average AOT values in the second half of the working week, usually from Wednesday to Friday or Saturday, and

Table 1. List of AERONET sun photometer stations and available data in alphabetical order.

AERONET Station	Coordinates	Measurement Period	Number of Days with Measurements
Carpentras	44.08 N 5.06 E	18.02.2003–13.04.2006	722
Fontainebleau	48.41 N 2.68 E	25.03.2002–30.11.2004	427
Hamburg	53.57 N 9.97 E	15.06.2000–25.10.2006	614
IFT-Leipzig	51.35 N 12.44 E	20.05.2001–14.03.2006	644
ISDGM_CNR	45.44 N 12.33 E	12.03.2002–08.11.2004	659
Ispra	45.80 N 8.63 E	28.06.1997–24.04.2006	1910
Karlsruhe	49.09 N 8.43 E	23.03.2005–31.10.2005	133
Laegeren	47.48 N 8.35 E	13.08.2003–05.07.2005	275
Mainz	50.00 N 8.30 E	05.11.2003–05.07.2006	360
Munich_Maisach	48.21 N 11.26 E	13.03.2002–09.06.2004	210
Oostende	51.23 N 2.93 E	05.09.2001–04.10.2003	296
Palaiseau	48.70 N 2.21 E	24.07.1999–03.10.2005	701
Paris	48.87 N 2.33 E	18.07.2000–24.11.2005	182
Venice	45.31 N 12.51 E	16.06.1999–01.04.2006	1586

Table 2. Average AOT440 (and AOT500 in brackets where available), standard deviation of AOT440, 95% confidence interval of average AOT440, average AOT440 weekday 95% confidence interval, and minimum (from Saturday until Tuesday)/maximum (from Wednesday until Saturday) average weekday AOT440 and corresponding weekday with result of t-test (in brackets) for the respective two weekdays of the AERONET sun photometer station data. Note that some of the stations do not measure AOT at a wavelength of 500 nm.

AERONET Station	Average AOT440 (AOT500)	St. Dev. (440)	CI 95% (440)	Average weekday CI 95% (440)	Min/Max AOT440 and corresponding weekday, significant (95%, 90%, no)
Carpentras	0.217 (0.174)	0.136	±0.009	±0.026	0.205 Sun/0.230 Wed (no)
Fontainebleau	0.279 (0.243)	0.173	±0.016	±0.042	0.236 Sat/0.331 Fri (90%)
Hamburg	0.233 (0.193)	0.175	±0.014	±0.036	0.207 Mon/0.251 Sat (90%)
IFT-Leipzig	0.281 (0.238)	0.186	±0.014	±0.037	0.252 Sun/0.315 Fri (95%)
ISDGM_CNR	0.390 (0.335)	0.260	±0.020	±0.053	0.350 Sun/0.429 Wed (95%)
Ispra	0.388 (0.317)	0.326	±0.015	±0.039	0.347 Tue/0.411 Sat (95%)
Karlsruhe	0.297 (0.251)	0.170	±0.029	±0.074	0.226 Mon/0.328 Sat (90%)
Laegeren	0.199 (0.166)	0.118	±0.014	±0.036	0.171 Sun/0.234 Thu (95%)
Mainz	0.262 (0.223)	0.156	±0.016	±0.042	0.230 Mon/0.311 Thu (95%)
Munich_Maisach	0.288 (-)	0.140	±0.019	±0.049	0.253 Mon/0.334 Sat (95%)
Oostende	0.281 (0.245)	0.214	±0.024	±0.064	0.254 Sun/0.323 Wed (no)
Palaiseau	0.248 (-)	0.158	±0.012	±0.030	0.217 Sun/0.276 Fri (95%)
Paris	0.265 (-)	0.164	±0.024	±0.061	0.222 Mon/0.289 Fri (90%)
Venice	0.338 (0.289)	0.234	±0.012	±0.030	0.314 Mon/0.360 Thu (95%)

minimum values on Sunday and Monday. The average over all 5 German stations also shows this cycle with a range of almost 20%. This average has been weighted with the number of measurement days of the single stations. The error bars denote the corresponding 95% confidence level. Additionally, we carried out two-tailed t-tests for the data of each station and compared minimum AOT on Monday with maximum AOT that occurred on Thursday, Friday or Saturday. We found the difference to be significant on the 95% level ($\alpha<0.05$) for the stations IFT Leipzig, Mainz and Munich-Maisach, and significant on the 90% level ($\alpha<0.10$) for Karlsruhe and Hamburg.

Figure 3 illustrates the mean relative weekly variability of AOT at 440 nm for the 3 AERONET stations situated in Greater Paris and their average as the relative percent departure from the mean value. Again, there is a clear weekly periodicity with a maximum AOT at the end of the working week that is greater than the minimum by about 20% which occurs on Sunday and Monday. The t-test for data of Monday and Friday resulted in a significant difference on the 95% level

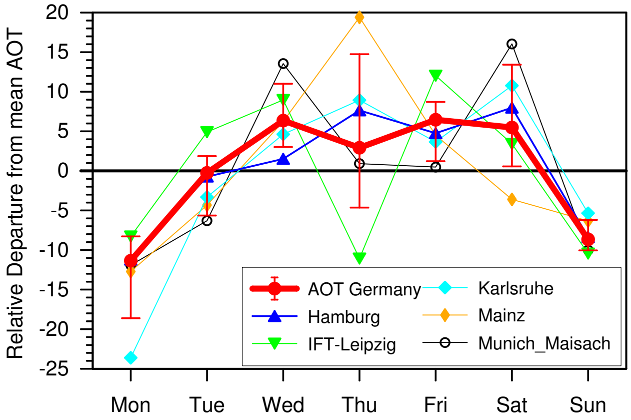

Fig. 2. Mean relative weekly variability of AOT at a wavelength of 440 nm for the 5 German AERONET stations and their average AOT at 440 nm computed as relative percent departure from mean value.

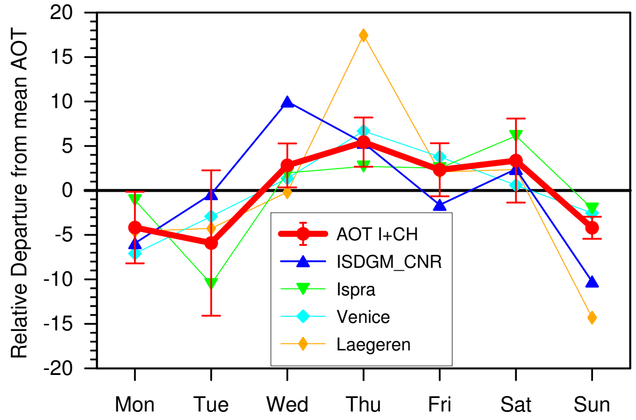

Fig. 4. Same as Fig. 2, but for 4 AERONET stations situated in Northern Italy and Switzerland. Note: The station "ISDGM_CNR" is situated in the city of Venice.

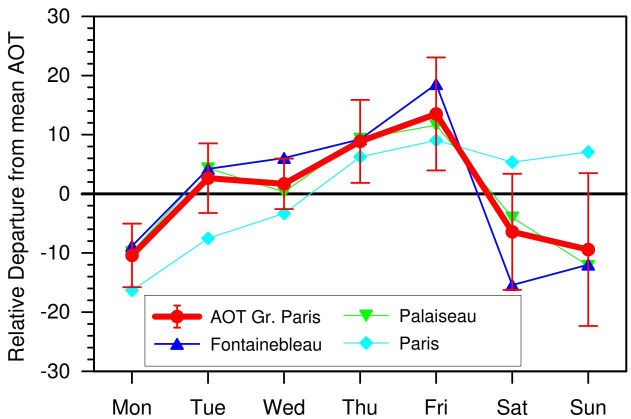

Fig. 3. Same as Fig. 2, but for the 3 AERONET stations situated in Greater Paris and their average.

for the stations Fontainebleau and Palaiseau, and a significant difference on the 90% level for Paris. In contrast to Germany, the decrease takes place already on Saturday, whereas the AOT over Germany is constantly high on Saturday. This is probably a consequence of less up-stream sources close to Paris in respect to prevailing westerly winds in comparison to the conditions in Germany, which leads to an advection of relatively clean air within a shorter time period. Note that the time series of the stations Fontainebleau (427 days) and Palaiseau (701 days) are much longer than the time series of the station Paris (182). This short time series might explain the deviant pattern observed at the station Paris that does not show a clear AOT decrease during the weekend but on Monday.

Figure 4 shows the mean relative weekly variability of AOT at 440 nm for 4 AERONET stations situated in Northern Italy and Switzerland and their average AOT at 440 nm, again as relative percent departure from the mean value. There is also a clear weekly cycle visible, although the difference between minimum and maximum is about 10% and, consequently, lower than in Germany or Greater Paris. The t-test for the data of single stations was carried out as comparison of the day with the minimum AOT in the period Sunday to Tuesday, and of the day with the maximum AOT in the period Wednesday to Saturday. For each of these four stations this resulted in a significant difference on the 95% level.

Data of the stations Oostende at the Belgian North Sea coast and Carpentras in Southern France do not display a significant weekly cycle, but a weak minimum on Sunday (Table 2). At both stations, the maximum occurred on Wednesday. The t-test resulted in $\alpha > 0.10$ for the comparison of data of Wednesday and Sunday, i.e. there is no significant difference between weekdays on the 90% level.

To address the question whether small differences in cloud cover in the course of the week as shown in Bäumer and Vogel (2007) could negatively influence the quality of the retrieved AOT, we additionally checked the number of days with measurements as a function of weekday. Summing the data of all the stations listed in Table 1 yields 8715 measurement days with a maximum on Friday (1268, +1.85%) and a minimum on Saturday (1224, −1.69%). The low percentage deviations from the mean do not argue for a remarkable influence on AOT retrieval, nor does the occurrence on Friday and Saturday. Since every single mean daily AOT value is derived from many single measurements on that day, we also carried out the same check for these single measurements summed up over all station data. 203 647 measurements were carried out in total with a maximum on Monday (29 611, +1.78%) and a minimum on Saturday (27 655, −4.94%). Again, these deviations are much smaller than the AOT anomalies, and therefore the possible influence on the AOT retrieval is supposed to be rather small. Nevertheless, they are consistent with the higher sunshine duration at the beginning of the week in comparison to Saturday as reported in Bäumer and Vogel (2007). Besides, we only applied level 2.0 data that are cloud-screened and quality-assured to minimize a possible negative impact of clouds on the AOT retrieval.

To check the robustness of the observed weekly AOT cycle, we repeated the comparison of maximum and minimum weekday AOTs as given in Table 2 but under the additional condition that only days of individual weeks are compared as suggested by Forster and Solomon (2003). Since the retrieval of AOT requires an undisturbed view to the sun which does not happen every day, this additional condition enormously reduces the data by 53% in total and therefore the statistical confidence accordingly. Nevertheless, at all stations still a similarly large difference between AOTs on maximum and minimum weekday is analyzed as in the complete dataset, but then the statistical significance is below 90% at the stations IFT-Leipzig, ISDGM_CNR, Laegeren, and Palaiseau. To avoid such a strong data loss, we decided to work with the complete data in this paper.

Additionally, we checked whether the weekly cycle is consistent in time by separately analyzing the first half and the second half of each time series of all stations with at least 600 measurement days to ensure statistical confidence. Reducing the number of single measurements by 50% again led to an increase in scattering with more random deviations on single weekdays, but this analysis also corroborated the finding of a weekly cycle including reduced AOT on Sunday in both the first and the second half of the time series. Some stations with relatively long time series as IFT-Leipzig, ISDGM_CNR, Ispra, and Venice also show a decreasing trend in time. This indicates a decrease in aerosol load over Central Europe during the last decade, a widespread phenomenon that is also known as brightening and follows a period of dimming (Wild et al., 2005). Similarly, we divided the time series in summer (March until August) and winter (September until February). A finer classification would have resulted in too small data subsets. All stations with sufficiently long time series and weekly cycles based on the complete dataset also showed weekly cycles both in winter and in summer. Again the statistical confidence is reduced in comparison to the complete data. At all stations, the average AOT in winter is significantly lower than in summer which is probably a consequence of the weaker photochemistry in winter. In Fig. 5, a comparison of the mean summer and winter relative weekly cycle of three selected stations Hamburg, Palaiseau, and Venice is shown. The range between maximum and minimum weekday AOT is higher in winter than in summer at all the three stations. Besides, the increase in the beginning of the week seems to be faster in summer than in winter at Hamburg and Palaiseau. We think that the photochemical formation of particles might be more important in summer than in winter which leads to an additional rapid relative increase in the beginning of the week in summer. Nevertheless,

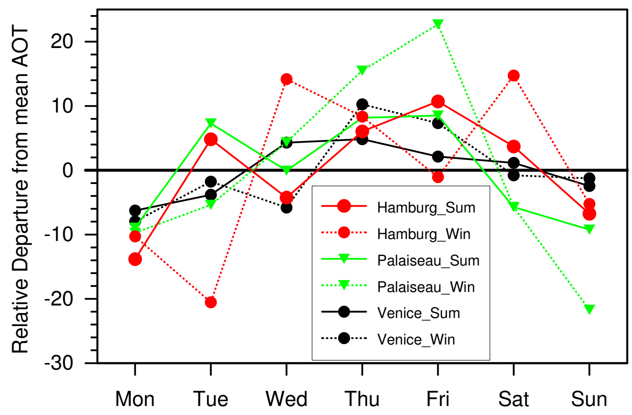

Fig. 5. Mean summer and winter relative weekly variability of AOT at a wavelength of 440 nm for the three selected AERONET stations Hamburg, Palaiseau, and Venice computed as relative percent departure from mean summer and mean winter value, respectively.

the lower (and more stable) boundary layer of winter leads to an increased range in the course of the week in winter. Additional particle emissions from heating might contribute to this phenomenon. At the end of the working week, a decrease in AOT is visible at all stations and seasons, except for Hamburg in winter that shows a relatively noisy weekly cycle. All graphs depict a further clear decrease on Sunday which continues until Monday, except for Palaiseau in winter. We conclude that the mean aerosol lifetime is at least one or two days on average. From the slower decrease on Saturday in summer in comparison to winter one could suspect continued formation processes, but additionally an average lower wind speed in summer causes a larger transportation timescale. The formation timescale we would estimate as of the order of one or two days from these results.

Since the anthropogenic aerosol fraction is supposed to be predominantly fine aerosol, we also analyzed the data subsets for fine and coarse aerosol as provided by AERONET separately. Since this partitioning is additionally based on clear sky measurements, the size of these data subsets with respect to single measurements is considerably reduced by about 90% relative to the pure AOT measurements based on direct sun measurements only. We found that the AOT is totally dominated by the fine fraction which accounts for 93%, whereas the coarse mode only contributes 7%. Among the stations given in Table 1, Oostende is the station with the highest coarse mode fraction that is 10%. Consequently, always the fine mode fraction generates the weekly cycles observed in the AOT of most of the stations. At some stations the coarse mode contributes to the weekly AOT cycle to a small amount. This can be either be caused by anthropogenic activities that generate relatively large particles as e.g. abrasion from brake pads and pavements, or dust from construction sites, or the AERONET retrieval technique does not completely separate the fractions.

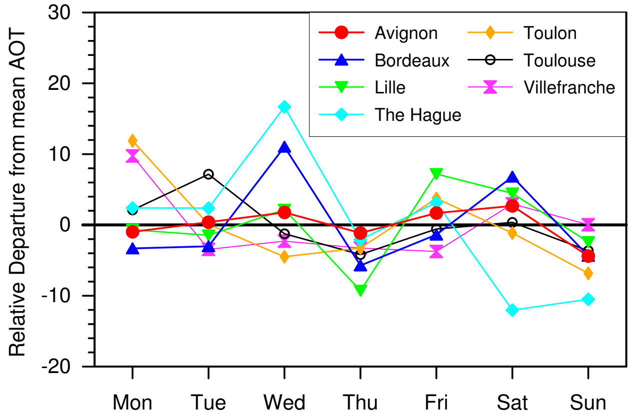

Fig. 6. Mean relative weekly variability of AOT at a wavelength of 440 nm for seven further AERONET stations situated close to the edges of the investigated area computed as relative percent departure from mean value.

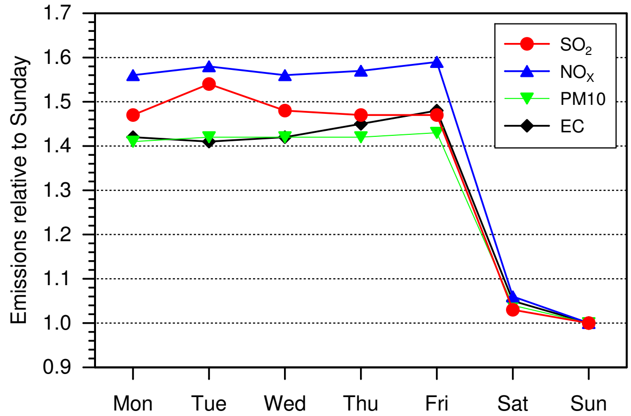

Fig. 7. Emissions of different gaseous and particulate substances by day of the week relative to Sunday for the domain 46° N 6° E to 51° N 10° E (South-Western Germany and adjacent areas) for June 2000.

To check if there could be negative effects of sun photometers breaking down e.g. during the weekend, we also analyzed the temporal distribution of the single measurements within different weekdays. Summing all station data, there are more single measurements on Sunday (29 128) than on Saturday (27 655). Since all data are quality-checked, this clearly does not argue for a maintenance impact which probably would lead to a decrease in the number of measurements during the weekend. The ratio between the number of measurements in the first half of the day before noon and the number of measurements in the second half of the day after noon is 1.02 on Saturday and 1.00 on Sunday. These are in the range of the ratios 0.99 to 1.03 that occur between Monday and Friday. Again, this does not argue for a weekend effect that might be caused by a maintenance effect. Furthermore, it demonstrates that for the average of our stations the weekly cycle is larger than the daily variability.

There are some further stations close to the edges of the investigated area which we subsequently have checked for weekly periodicities too. Figure 6 gives the weekly variability of this second set of stations. Some stations in Southern France (Toulouse, Toulon, Avignon, Bordeaux, Villefranche) do not display significant weekly cycles. Similarly, close to the western edge of the area, Lille does not have a significant weekly AOT cycle, and The Hague displays one but with large scattering from day to day. Nevertheless, at some stations a minimum around Sunday is visible (Avignon, The Hague), and none of the stations show above-average values on Sunday.

4 Conclusions

Many stations as all the German stations, the stations in or close to Paris, France, or the stations in or close to Venice, Italy, show significant weekly periodicities of the aerosol optical thickness. Low AOT values on Sunday and Monday are in contrast to relatively high AOT values from Wednesday to Friday or Saturday. To our knowledge, this is the first time that ground-based measurements with high accuracy demonstrate these weekly AOT cycles at many stations in an area with a considerable horizontal extent. In comparison to the characteristic ranges of the diurnal AOT variability shown by Smirnov et al. (2002) but for other stations, the weekly variability is at least of the same order as the mean daily variability at various urban or industrial sites. At the stations analyzed in this study, the weekly AOT cycle is larger than the daily variability according to our results. From a further analysis of data from stations close to the edges of the investigated domain, we follow that the area in which significant weekly AOT periodicities as derived from AERONET are widespread in Europe might be limited to Central Europe.

These weekly periodicities are a result of human activity that leads to a characteristic weekly emission cycle. In Fig. 7, typical relative weekly emission cycles of gaseous and particulate substances for South-Western Germany and adjacent areas are shown that consist of nearly constant emissions from Monday to Friday and distinctly lower emissions on Saturday and Sunday (Th. Pregger, Institut für Energiewirtschaft und Rationelle Energieanwendung (IER), University Stuttgart, personal communication, 2006). The applied emission model is based on work carried out in the EUROTRAC subproject GENEMIS (Friedrich and Reis, 2004). Emissions on working days are on average 40% to 60% higher than emissions on Sunday or Saturday. A very similar weekly emission cycle for NO_x for Germany can be found in Beirle et al. (2003). This means that there is a phase shift of about one day at the end of the week between the emission cycle and the AOT cycle in Germany and Northern Italy. This is probably caused by the typical time scale for the advection of cleaner air. In Greater Paris, the advection of cleaner air seems to take place already on Saturday predominantly, so that there is no phase shift between the emission cycle and the AOT cycle. Similarly, in the beginning of the week a phase shift occurs with respect to the increase in emissions and AOT. A possible explanation for that is that not all the aerosol particles are directly emitted, but are formed in the atmosphere from gaseous precursors. In Germany, it takes until Wednesday to reach the maximum AOT level that is almost constant until Saturday. This suggests the conclusion that the formation of secondary aerosols typically takes about two days in Germany on average, since the emissions are as high on Monday as during Tuesday until Friday. All the single stations in Germany seem to show a clear increase from Monday until Wednesday in the same manner. A little different from that, in Greater Paris a more constant increase from Monday until Friday is visible in the AOT, which makes it difficult to derive an approximated time scale for secondary aerosol formation. In Italy, the situation is similar to Germany, with a plateau of relatively high AOT from Wednesday until Saturday (Fig. 4), which also argues for a formation time scale of about two days. The average lifetime of aerosol can be estimated as at least two days from our results.

By applying checks with respect to the temporal distribution of the data among the weekdays, we could exclude that the weekly cycles were caused by a maintenance effect. We also do not think that slightly different retrieval conditions as a consequence of a weekly cycle in cloud cover could have caused artificial weekly cycles in AOT.

We conclude that, in Germany, there is a period from Wednesday to Saturday with a relatively strong anthropogenic direct aerosol effect, and a weaker one during the rest of the time. In Germany, a widespread weekly temperature periodicity with relative maxima on Wednesday and relative minima on Saturday has been observed recently (Bäumer and Vogel, 2007). Hence we follow that the relatively strong anthropogenic direct aerosol effect indicated by relatively high AOT may contribute to the observed weekly temperature periodicity.

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